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This project utilizes key empirical principles based on known high-temperature superconductors in order to design and fabricate novel heterostructures that exhibit high-temperature superconductivity. Design of heterostructures is accomplished using the predictive capabilities of ab initio density functional theory (DFT) calculations combined with many-body theories of electronic structure, such as dynamical mean field theory (DMFT). Fabrication is accomplished with atomic-layer control by oxide molecular beam epitaxy (MBE) synthesis. Finally, the theoretically derived physical and electronic structures are tested using a variety of microscopic and macroscopic

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ABSTRACT

This project utilizes key empirical principles based on known high-temperature superconductors in order to design and fabricate novel heterostructures that exhibit high-temperature superconductivity. Design of heterostructures is accomplished using the predictive capabilities of ab initio density functional theory (DFT) calculations combined with many-body theories of electronic structure, such as dynamical mean field theory (DMFT). Fabrication is accomplished with atomic-layer control by oxide molecular beam epitaxy (MBE) synthesis. Finally, the theoretically derived physical and electronic structures are tested using a variety of microscopic and macroscopic characterization techniques. Our approach to the design of the heterostructures is based on emulating key common properties exhibited by the cuprate family of high-temperature superconductors.

Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received		Paper
09/09/2013	3.00	Hanghui Chen, Divine P. Kumah, Ankit S. Disa, Frederick J. Walker, Charles H. Ahn, Sohrab Ismail-Beigi. Modifying the Electronic Orbitals of Nickelate Heterostructures via Structural Distortions, Physical Review Letters, (05 2013): 186402. doi: 10.1103/PhysRevLett.110.186402
09/09/2013	4.00	A. S. Disa, D. P. Kumah, J. H. Ngai, E. D. Specht, D. A. Arena, F. J. Walker, C. H. Ahn. Phase diagram of compressively strained nickelate thin films, Applied Physics Letters Materials , (09 2013): 0. doi:
10/23/2014	5.00	Sohrab Ismail-Beigi, Frederick J. Walker, Charles H. Ahn, Divine P. Kumah, Ankit S. Disa, Joseph H. Ngai, Hanghui Chen, Andrei Malashevich, James W. Reiner. Tuning the Structure of Nickelates to Achieve Two-Dimensional Electron Conduction, Advanced Materials, (03 2014): 0. doi: 10.1002/adma.201304256
12/11/2012	1.00	Karyn Le Hur, Chung-Hou Chung, I. Paul. Designing heterostructures with higher-temperature superconductivity, Physical Review B, (07 2011): 24526. doi: 10.1103/PhysRevB.84.024526
12/11/2012	2.00	J. Kirtley, B. Kalisky, J. Bert, C. Bell, M. Kim, Y. Hikita, H. Hwang, J. Ngai, Y. Segal, F. Walker, C. Ahn, K. Moler. Scanning SQUID susceptometry of a paramagnetic superconductor, Physical Review B, (06 2012): 224518. doi: 10.1103/PhysRevB.85.224518
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TOTAL:

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(c) Presentations

D. P. Kumah (contributed), 2014 American Physical Society March Meeting, Denver, CO, "A structural route to tuning the orbital structure of nickelates," (2014).

D. P. Kumah (contributed), 2013 Materials Research Society Fall Meeting, Boston, MA, "Resonant x-ray studies of NdNiO3 heterostructures," (2013).

D. P. Kumah (invited), George Mason University, Physics Seminar, Fairfax, VA, "Correlating atomic structure and emergent phenomena at complex oxide interfaces," (2014).

A. S. Disa (contributed), American Physical Society March Meeting, Presentation, March 2014, Denver, CO, “Renormalization of the nickelate phase diagram in strained thin films," (2014).

A. Malashevich (contributed), 2013 Materials Research Society Fall Meeting, Boston, MA, "Surface Structure, Screening Effects, and Electronic States of LaNiO3 Thin Films from First Principles," (2013).

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Technology Transfer

Engineering Superconductors by Design – final report

C.H. Ahn, S. Ismail-Beigi, K.A. Moler, F.J. Walker

This project utilizes key empirical principles based on known high-temperature superconductors in order to design and fabricate novel heterostructures that exhibit high-temperature superconductivity. Design of heterostructures is accomplished using the predictive capabilities of *ab initio* density functional theory (DFT) calculations combined with many-body theories of electronic structure, such as dynamical mean field theory (DMFT). Fabrication is accomplished with atomic-layer control by oxide molecular beam epitaxy (MBE) synthesis. Finally, the theoretically derived physical and electronic structures are tested using a variety of microscopic and macroscopic characterization techniques. Our approach to the design of the heterostructures is based on emulating key common properties exhibited by the cuprate family of high-temperature superconductors(*1*). Specifically, there are the four critical features of the these materials that we set out to engineer:

1. Spin ½ system
2. Two-dimensional conduction
3. Strong antiferromagnetic correlations
4. Absence of orbital degeneracy

We use as a parent material for this project the rare-earth nickelates ($RNiO_3$, R = La, Nd), due to their similarity with the cuprates (Ni is adjacent to Cu in the periodic table). In the bulk, this family of oxides exhibits spin-½ physics and a phase diagram with a tunable antiferromagnetic ground state. Furthermore, many-body theoretical analysis indicates that superconductivity in this system could occur at temperatures higher than in the cuprates. However, the bulk nickelates are three-dimensional conductors and possess an orbitally degenerate electron configuration. The goal of our work is to engineer all four of the above criteria into a single artificial heterostructure and experimentally test for signs of superconductivity.

Two-dimensional conduction in $LaNiO_3$

In bulk nickelates, the electrical transport is three-dimensional due to the symmetry of the material. To realize the criterion of two-dimensional conduction in nickelate materials, the most direct approach is to constrain the conduction in two dimensions by growing a film with a thickness close to a unit cell. However, it is experimentally observed that thin nickelate films undergo a transition to an insulating state below a certain critical thickness. For $LaNiO_3$, the most conducting of the rare-earth nickelate family, a metal-insulator transition occurs below a thickness of ~5 unit cells, limiting the ability to achieve two-dimensional conduction.

In our research, we were able to circumvent this limitation by understanding the microscopic origin of the metal-insulator transition. We combined x-ray diffraction measurements with DFT calculations on $LaNiO_3$ thin films to discover that the conducting NiO_2 planes are made insulating due to distortions that occur at the surface of the film (*2*). The distortions cause a buckling of the Ni-O bond angle, and hence a

reduction in the Ni-O orbital hybridization responsible for the conduction. As a consequence, we deduced that removing the vacuum surface and replacing it with an insulating capping layer should remove the distortions and result in metallic conduction in films below the observed critical thickness. Indeed, we found that $LaNiO_3$ films as thin as 3 unit cells thick exhibit metallic behavior down to low temperatures when capped with 6 unit cells of insulating $LaAlO_3$, and that the topmost NiO_2 planes of the film are flat and bulk-like (Fig. 1). We also fabricated superlattice samples with multiple repeating $LaNiO_3$ and $LaAlO_3$ layers that show structural and electrical characteristics akin to those in the capped film.

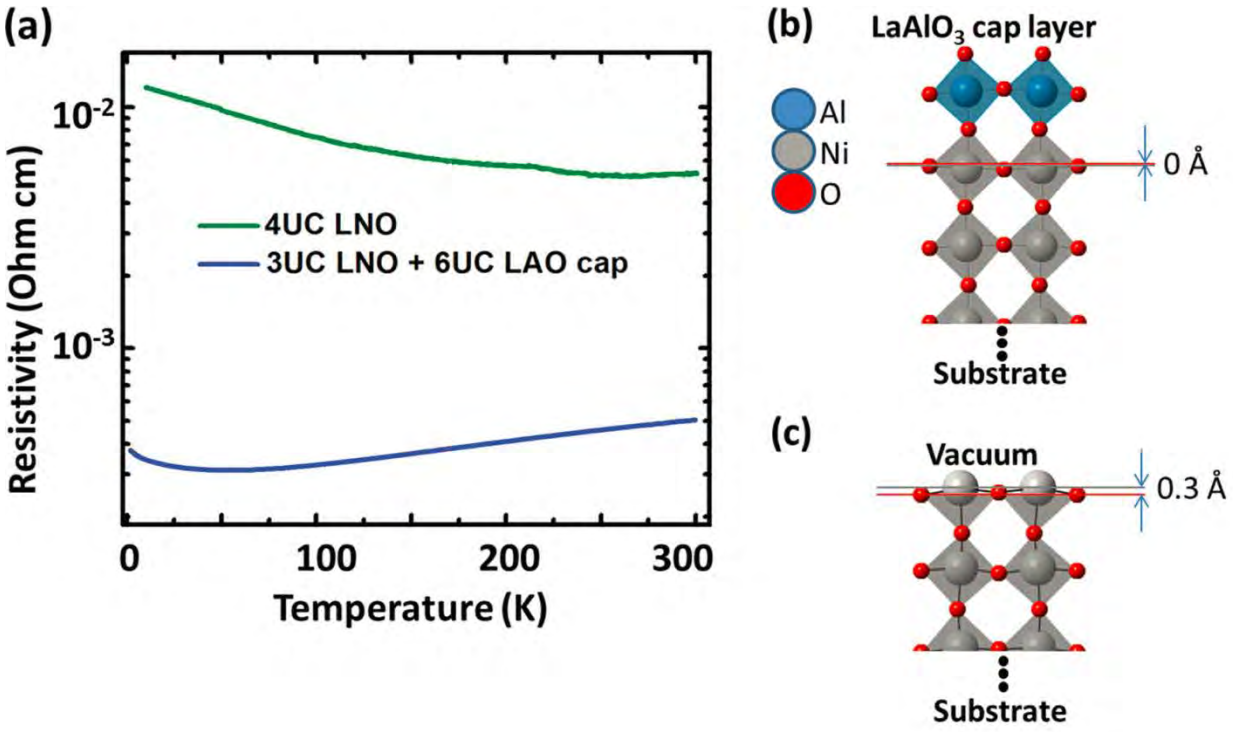

Figure 1. (a) Electrical resistivity for thin films of $LaNiO_3$ both uncapped and capped with $LaAlO_3$, (b) the experimentally determined atomic structure near the surface of the $LaNiO_3$ for a capped film and for (c) an uncapped film.

Importantly, we used macroscopic transport measurements at low temperatures and high magnetic fields to confirm the two-dimensional nature of the conduction. In particular, the low temperature (< 25K) transport exhibits weak localization, whereby the zero-field resistivity increases as the temperature is lowered and negative magnetoresistance is observed. This behavior in a metal is a hallmark of two-dimensional electronic conduction, where the field and temperature dependence characterize the electron inelastic mean free path, which is roughly 10 nm for 3 uc-thick $LaNiO_3$, significantly larger than the thickness of the film. Thus, we have been able to achieve two-dimensional metallic conduction in nickelates.

$LaNiO_3$-$NdNiO_3$

The requirement of strong antiferromagnetic correlations is evidenced in bulk nickelates by the electronic phase diagram, in which transitions between a high-temperature paramagnetic metal and a low-temperature antiferromagnetic insulator can be tuned by changing the rare-earth ion. From the standpoint of realizing high-Tc-like behavior in nickelate heterostructures, the tunability of magnetic and electronic correlations must be studied for thin films of nickelates. Thus, we explored the phase diagram of epitaxially grown nickelate thin films upon which our heterostructures are based (*3*).

Specifically, we mapped out the region of the phase diagram between Nd and La for compressively strained 8 unit cell thick films. We mapped out the phase diagram by growing solid solutions of $LaNiO_3$ and $NdNiO_3$ on $LaAlO_3$ substrates by molecular beam epitaxy, varying the rare-earth composition of $Nd_{1-y}La_yNiO_3$ in the range $0 \leq y \leq 1$. The resistivity of these films was measured as a function of temperature to identify the metal-insulator transition temperature. By plotting the metal-insulator transition temperature of the thin films as a function of the average ionic radius, we determined that the thin film phase diagram has a similar shape to the bulk phase diagram, but the transitions are renormalized to lower temperatures (Fig. 2). That is, for a given composition in the range studied, the metal-insulator transition temperature is roughly 50K lower for the thin film than for the equivalent bulk material.

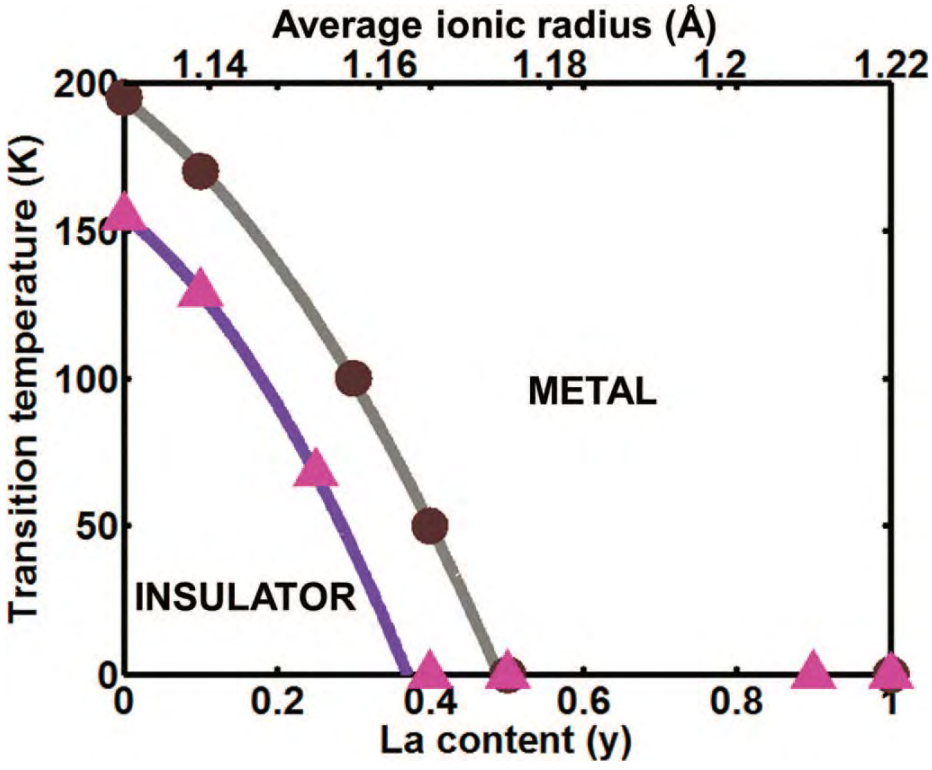

Figure 2. Phase diagram of $Nd_{1-y}La_yNiO_3$ for bulk (brown circles, grey line) and 8 unit cell thick thin films grown on $LaAlO_3$ (pink triangles, purple line).

The origin of this renormalization could arise due to a number of different factors, including oxygen vacancies, reduced dimensionality, and epitaxial strain. We ensured a low concentration of oxygen vacancies in our thin films by growing them in an activated oxygen plasma environment and post-

annealing the films in 1 atm. of O_2; the oxygen stoichiometry is confirmed by x-ray absorption (XAS) measurements. To test the effects of reduced dimensionality, we measured the resistivity of $NdNiO_3$ films of different thicknesses. We found that even bulk-like films with thicknesses of ~20nm grown on $LaAlO_3$ show the same metal-insulator transition temperature as an 8 unit cell thick film. This result rules out dimensionality as a driving force for the lower transition temperature observed in thin nickelate films and points to epitaxial strain as the chief source of this phenomenon. This hypothesis was corroborated by x-ray diffraction, which showed that the compressively strained thin films exhibit Ni-O bond lengths and bond angles similar to bulk samples under hydrostatic pressure. Further, x-ray spectroscopy shows a greater overlap between Ni and O orbitals due to strain, which correlates with the metal-insulator transition temperature and explains the observed phase diagram renormalization.

Scanning SQUID susceptometry of $LaNiO_3$ films

In addition to designing and fabricating novel materials, we contributed to developing a reliable tool to characterize potential novel quantum behavior in such systems. Spatially averaged transport and magnetic measurements survey only macroscopic regions of a sample. Such measurements may not be sensitive to microscopic regions of the sample that exist in interesting magnetic or superconducting states, or may not be appropriate if co-existence of such phases occurs on microscopic length scales. We performed scanning superconducting quantum interference device (SQUID) susceptometry on $LaNiO_3$ films (*4*). These experiments combined measurements of the local magnetization and susceptibility with derived analytical expressions of the magnetic response for paramagnetic and superconducting materials. This technique can be used to detect, with high spatial resolution and sensitivity, the concentrations of spins and/or superconducting carriers. Even in the $LaNiO_3$ films with small paramagnetic susceptibility, spatially resolved measurements of the spin concentration could be obtained with high contrast (Fig. 3).

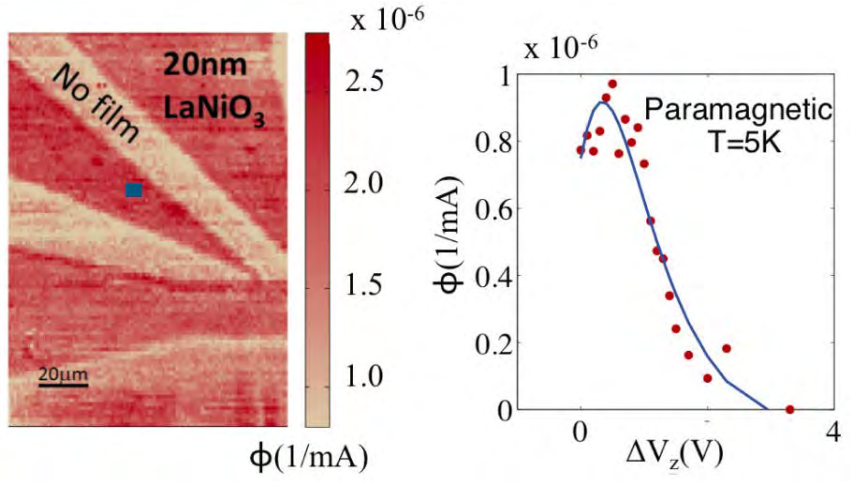

Figure 3. (left) Scanning SQUID susceptibility image of a patterned 20nm thin film of $LaNiO_3$, and (right) the susceptibility approach curve at the position of the square symbol on the left image, including data (red dots) and theoretical fit (blue line).

Designing high-Tc-like heterostructures

The ground state electronic configuration in the bulk nickelates is d^7 with 6 electrons filling the 3 t_{2g} orbitals and the remaining electron being shared by the e_g orbitals (*3d* orbitals with x^2-y^2 and $3z^2$-r^2 symmetry). This degenerate configuration arises due to the essentially cubic symmetry of the underlying crystal. In order to lift the orbital degeneracy, the orbital energies must be separated by altering the symmetry artificially.

One possible way of doing this would be to layer thin nickelate layers with insulating barriers in a repeating superlattice geometry, as predicted by Chaloupka and Khaliullin *(1)*. Indeed, we experimentally showed that two-dimensional conduction can be achieved in such structures with alternating $LaNiO_3$ and insulating $LaAlO_3$ layers. However, both theoretically and experimentally we find that such a heterostructure fails to appreciably remove the orbital degeneracy. The problem here lies in the fact that while the insulating barrier layers reduce the out-of-plane hopping compared to the in-plane hopping (which facilitates the two-dimensional conduction), the environment around each Ni ion remains essentially bulk-like. Thus, the width of the energy band derived from the $3z^2$-r^2 orbital is reduced, but the average energy of the band remains the same as that of the band derived from the x^2-y^2 orbital. Hence, the two bands remain degenerate.

We had previously found that the ions near the surface of the thin films experience large distortions. These distortions are driven by the polar electric field that builds due to the alternating charged NiO_2^- and LaO^+ planes, and by the fact that at the vacuum-surface interface, the inversion symmetry is broken. The result is that the out-of-plane apical Ni-O bonds are broken and elongated relative to the in-plane ones. DFT calculations of the atomic structure agree well with the atomic structure determined using x-ray diffraction experiments. From the DFT calculations, we derive the electronic structure and find that for the topmost layer in the film, which undergoes the greatest distortion, the energy splitting between the $3z^2$-r^2 and x^2-y^2 orbitals is large, 1.29 eV. Combined with the $3z^2$-r^2 band narrowing due to reduced out-of-plane hopping into the vacuum, this large splitting lifts the degeneracy.

Using this observation from the thin film case, we designed a robust, three-dimensional material that should reproduce high-Tc-like behavior in the nickelates (*5*). The heterostructure is designed with large built-in electric fields using inversion symmetry breaking in a three-component superlattice. This geometry mimics the physical mechanism that drives Ni-O bond asymmetries, and hence orbital polarization, in thin films. The superlattice is composed of layers of $LaNiO_3$ sandwiched between $LaTiO_3$ and an insulating barrier. Due to the relative electronegativity of Ni and Ti, the $LaTiO_3$ transfers an electron to the $LaNiO_3$ layer. A polar charge distribution thus develops, which creates electric fields akin to those observed at the surface of $LaNiO_3$ thin films (Fig. 4). This field alternates in the superlattice, and due to broken inversion symmetry, points towards the Ni. As a result, the apical Ni-O bonds are elongated leading to a lifting of the orbital degeneracy.

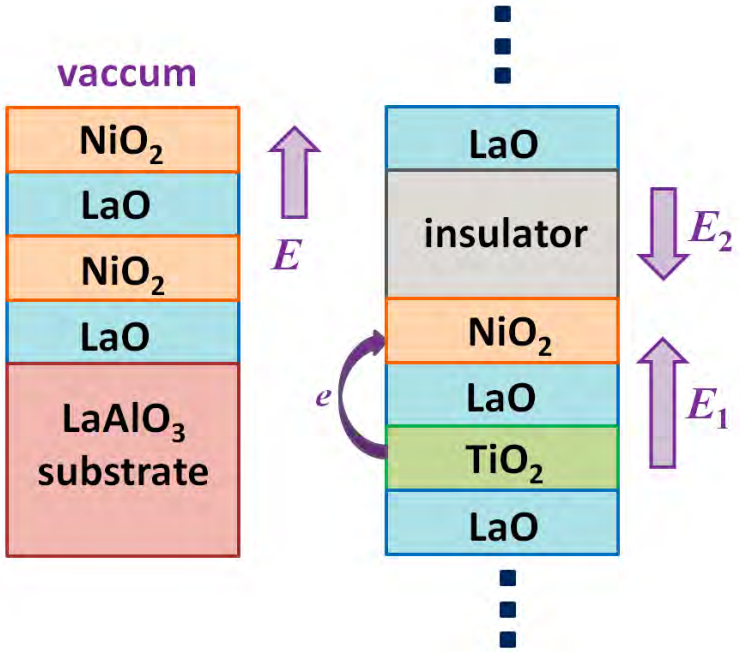

Figure 4. Schematic diagram of layer sequence and internal fields for (left) a thin film of $LaNiO_3$ and (right) a three-component superlattice.

We examined in detail one instance of the superlattice with RbF as the insulator (*i.e.* a $(LaTiO_3)_1/(LaNiO_3)_1/(RbF)_2$ superlattice). We find that, structurally, the environment around the NiO_2 layer resembles that experienced near the vacuum-surface interface of the thin film. Further, the density of states projected onto the e_g orbitals also matches that of the thin film surface with a narrow $3z^2$-r^2 band split by 1.25 eV from the center of the x^2-y^2 band. A small amount of electron doping can split the bands even farther such that the orbital makeup of the Ni resembles that of the high-Tc superconductors, with the $3z^2$-r^2 band fully occupied and the Fermi level cutting only through the x^2-y^2 band, which is half occupied. The calculations show that a similar band structure exists for three-component superlattices with other insulating barriers as well, including $LaAlO_3$ and $SrTiO_3$.

Experimental tests of high-Tc -like behavior for three-component superlattices

Experimentally, we grew $LaTiO_3/LaNiO_3/(LaAlO_3)_3$ superlattices using an activated oxygen source and molecular beam epitaxy to confirm the theoretical model discussed above. Three-component superlattices with $LaAlO_3$ as the insulating layer (instead of RbF) are predicted to exhibit high-Tc-like behavior that could be used to generate superconductivity. Two experimentally accessible predictions we sought to validate are 1) the unidirectional transfer of electron charge from the Ti to the Ni, which dopes the Ni internally; and 2) a polar field with associated structural distortions pointing towards the nickelate layer.

First, to confirm the electron transfer from Ti to Ni, we used x-ray absorption spectroscopy to look at the valence states of the Ti ions in the three-component superlattice. Separately, the Ti and Ni ions in $LaTiO_3$ and $LaNiO_3$ are both in 3+ oxidation states. When combined into the superlattice, we expect a transfer

such that $Ti^{3+} \rightarrow Ti^{4+}$ and $Ni^{3+} \rightarrow Ni^{2+}$. We compared the Ti *L* edge spectrum of the three-component superlattice to those of bulk $SrTiO_3$ (with Ti^{4+}) and $LaTiO_3$ with (Ti^{3+}) and found that the Ti valence in the superlattice is predominantly Ti^{4+}. Hence, the transfer of an electron from the Ti to the Ni in the three-component structure was confirmed.

Second, the electron transfer is predicted to cause an electric field pattern in the superlattice, which should drive distortions of the ions from their equilibrium positions. These distortions can be evaluated by using high-resolution x-ray diffraction measurements to determine the atomic structure. Comparing the dipole moments in each layer, we found excellent agreement between experiment and theory. The polar field points towards the NiO_2 planes from both above and below, which is a key design element for mimicking high-Tc superconductivity.

Summary

The rare-earth nickelates have been targeted as a candidate to observe high-T_c superconducting behavior, akin to the cuprates due to the presence of spin-1/2, correlated, antiferromagnetic fluctuations suggested by the electronic phase diagram of the nickelates. We have predicted using dynamical mean field theory calculations high T_c superconductivity could be potentially achieved in nickelates due to the correlations and covalency present in this system. In order realize the specific electronic structure models used in these models, artificial heterostructures of nickelates must be engineered that exhibit two-dimensional conduction with high-Tc-like electronic structure, which are lacking in the bulk. Through our research, we discovered the fundamental physical mechanisms and practical design principles necessary to realize this goal.

Using x-ray diffraction and *ab initio* theory, we discovered that nickelate thin films undergo a metal-insulator transition below a critical thickness due to unscreened polar distortions at the vacuum-surface interface. Leveraging this understanding, we were able to engineer two-dimensional conduction in $LaNiO_3$ films as thin as 3 unit cells by replacing the vacuum-surface interface with an insulating capping layer. In addition, we utilized the tunable growth capabilities of molecular beam epitaxy to map out a phase diagram for nickelate thin films. In doing so, we demonstrated a renormalization of the bulk phase diagram to lower temperatures for thin films that is chiefly due to coherent epitaxial strain, rather than dimensional confinement. In the process of conducting this work, we discovered through DFT calculations that the surface Ni in $LaNiO_3$ thin films exhibits a distinct electronic structure as a result of distortions in the physical structure caused by a polar electric fields and inversion symmetry breaking. From this observation, we developed a three-component superlattice emulating the characteristics of the thin film surface, using charge transfer to drive internal polar fields and concomitant structural distortions. Our DFT calculations predict that these structures have cuprate-like behavior, which may be a candidate for developing new high-Tc-superconductors. We confirmed these predictions using x-ray diffraction and spectroscopy for an experimentally fabricated three-component superlattice that has a high-Tc-like electronic structure.

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Doi 10.1103/Physrevb.85.224518).
5. H. Chen, D. P. Kumah, A. S. Disa, F. J. Walker, C. H. Ahn, S. Ismail-Beigi, Modifying the Electronic Orbitals of Nickelate Heterostructures via Structural Distortions. *Physical Review Letters* **110**, 186402 (2013)10.1103/PhysRevLett.110.186402).